

Search for pick-up ion generated Na⁺ cyclotron waves at Mercury

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[1] Magnetometer data was examined from the two Mariner 10 passages through Mercury's magnetosphere for evidence of Na⁺ electromagnetic ion cyclotron waves (ICW). These waves are expected to be produced if the thermalization of newly created Na⁺ pick-up ions occurs. We found no evidence of Na⁺ ICW. The spacecraft's rapid passage through Mercury's magnetosphere meant that the ambient magnetic field was only relatively constant for at most $\sim 3-4$ Na⁺ ICW wave periods throughout each transversal, therefore limiting the spectral resolution of the Na⁺ cyclotron frequency. Our computations indicate that the wavelengths of Na⁺ ICW may be on the order of the system scale lengths. It is, therefore, questionable whether these waves can grow to sufficient amplitude to effectively thermalize these pick-up ions and incorporate them into Mercury's magnetosphere. Citation: Boardsen, S. A., and J. A. Slavin (2007), Search for pick-up ion generated Na⁺ cyclotron waves at Mercury, Geophys. Res. Lett., 34, L22106, doi:10.1029/2007GL031504.

1. Introduction

- [2] Mercury's Sodium exosphere was discovered by Earth-based observations [Potter and Morgan, 1985] and has been observed to form a tail in the anti-sunward direction due to Solar radiation pressure [Potter et al., 2002]. Due to a strong non-thermal energetic component [Potter and Morgan, 1997] there is a spatial overlap between this exosphere and the magnetosphere and magnetosheath (see review by Slavin [2004]) as illustrated in Figure 1. The exospheric Na that is not reabsorbed by Mercury's surface will eventually be ionized by solar radiation. Accordingly, the question arises as to whether Na⁺ pick-up ions make a significant contribution to the mass loading of Mercury's magnetosphere.
- [3] From analysis of the Na exosphere and Na⁺ tracing *Ip* [1986] and *Cheng et al.* [1987] suggested that Na⁺ could make up between 10% and 50% of the magnetosphere ion plasma composition. *Othmer et al.* [1999] by analysis of possible field line resonances placed a lower limit of 14% for the Na+ contribution. Thus there could be an important heavy ion component to Mercury's magnetosphere. If this is true, then rapid thermalization must take place in order to incorporate Na⁺ into Mercury's magnetospheric plasma population. Without this thermalization Na⁺ would be rapidly lost from the magnetosphere due to its large Larmor radii [*Delcourt et al.*, 2002, 2003]. Scattering of pick-up

ions due to wave-particle interactions is the principal mechanism for their thermalization at comets and planets [e.g., *Lee*, 1989; *Terasawa*, 1989].

[4] Figure 2a shows the ion gyro radii of different

initially "picked up" ion species versus perpendicular component of the bulk plasma flow velocity. These freshly created pick-up ions will form a ring distribution (assuming the scale lengths allow) which will be highly unstable to the generation of plasma waves [Lee, 1989; Tsurutani, 1991]. The primary mode excited by pick-up ions is the ICW [Thorne and Tsurutani, 1987]. ICW are waves whose wave vector is nearly aligned with the magnetic field for which the wave frequency resonates with the cyclotron frequency of an ion species, in this paper the resonant ion is Na⁺ pickup ion. The wave power lies largely in the transverse components of the wave field. The wave mode can be either Left (L) or Right (R) handed in the bulk plasma flow frame (i.e., the medium that supports the oscillations). In the bulk plasma flow frame the phase velocities of both the R and L mode waves are on the order of the Alfven velocity, but for the L mode the phase velocity drops to zero at the ion cyclotron resonances. It is critical that these waves when Doppler shifted to the pick-up ion frame of reference be left handed and in cyclotron resonance in the frame traveling with the Na⁺ pick-up ions. These waves will cause the Na⁺ ring distribution to rapidly diffuse in pitch angle followed by diffusion in energy (i.e., "thermalize") [Terasawa, 1989]. In this paper we will look for the presence of Na⁺ ICW in the Mariner 10 magnetometer data obtained during two transits of Mercury's magnetosphere. Simple estimates of the wavelengths will be derived and compared with the scale size of the Mercury system.

2. Data Analysis

- [5] The triaxial fluxgate magnetometer flown on Mariner 10 made 25 vector measurements per second, a Nyquist frequency of 12.5 Hz. The magnetometer ranges where ±128 nT and ±512 nT with digital resolutions of 0.26 nT and 1.0 nT respectively. The instrument noise level was in the range of 0.03–0.07 nT. High resolution magnetometer data for both flybys I and III where obtained from the Planetary Data System (PDS). The high resolution data for encounter III was recalibrated using 6s data published by *Lepping et al.* [1979].
- [6] Figure 2b shows the cyclotron frequency versus magnetic field magnitude for various ion species, the right ordinate is the minimum sampling time required to resolve the corresponding frequency. The Na^+ cyclotron frequency (f_{cNa+}) and minimum sampling times for different regions are indicated. Typically sampling times of a few minutes are required to resolve f_{cNa+} over which the ambient magnetic field has to be fairly steady.

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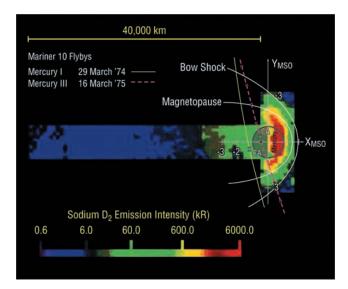


Figure 1. Earth based observations of the Mercury Na exosphere (taken from Figure 2 of *Potter et al.* [2002], by permission of the Meteoritical Society) are shown. Mariner 10 flybys 1 and 3, along with a model bowshock and magnetopause, in Mercury Solar Orbital (MSO) coordinates, are superimposed. This figure suggests that Mariner-10 was in a good position to detect wave signatures of Na pickup ions formed by photo-ionization of Na. CA indicates closest approach.

[7] Fourier spectra of the field magnitude, parallel, and perpendicular components were generated and examined for both flybys. The time interval for each spectrum was adjusted such that spectral peaks at and near the Na⁺

cyclotron frequency (f_{cNa+}) could be resolved. Time intervals for which a constant ambient magnetic field (i.e. cyclotron frequency) could not be reasonably assigned were discarded. Unfortunately, due to the rapid transversal of Mariner 10 through Mercury's geospace one can observe wave trains lasting at most only a few cycles over which the ambient field is fairly constant.

3. Observations

[8] Examination of the Mariner 10 data has yielded very few spectra with a spectral peak near the Na $^+$ cyclotron frequency (f_{cNa+}) , and in those cases no coherent wave train was present. Figure 3a shows a spectrum in which a peak was detected near f_{cNa+} during the first flyby. This spectrum was taken when Mariner 10 was inside the magnetosphere. A spectral peak near 0.04 Hz at \sim 1.2 f_{cNa+} is observed in all but one of the transverse components. The power in the parallel peak is close to that of the perpendicular peak which is not characteristic of parallel propagating ICW.

[9] Assuming that the waves are parallel to B, the Doppler shifted frequency observed in the spacecraft frame is dependent on the parallel component of the spacecraft velocity relative to that of the pick-up ion velocity divided by the wavelength. For this case the satellite velocity is \sim 5 km/s along the field line and the parallel component of the pick-up ions is at most a few km/s. An assumed wavelength of 220 km is estimated from the evaluation of equation (2), discussed later in this paper. In order to evaluate equation (2), assuming a proton background plasma, the electron density (N_e), Alfven velocity (V_A), and the parallel component of the flow velocity ($V_{F||}$) must be estimated. Using the measured values of 3 cm⁻³ for N_e [Ogilvie et al.,

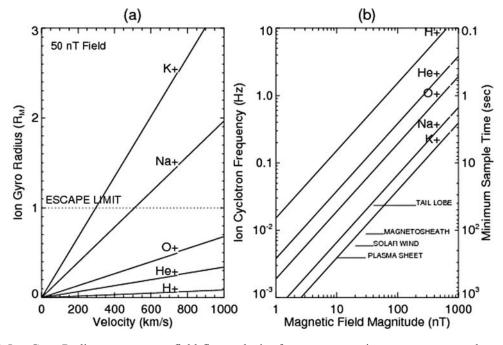


Figure 2. (a) Ion Gyro Radius versus cross field flow velocity for a representative outer magnetosphere magnetic field value [*Slavin*, 2004], the Na $^+$ gyro radius approaches the scale size of the magnetosphere around 500 km/s, marked as escape limit on the plot. One Mercury radii (1 R_M = 2439.7 km) was chosen because the planet is large compared to the size of its magnetosphere as illustrated in Figure 1. (b) Ion cyclotron frequency (left scale) and the required minimal dwell time (right scale) versus magnetic field strength. Representative field ranges for various regions in Mercury's magnetosphere are indicated.

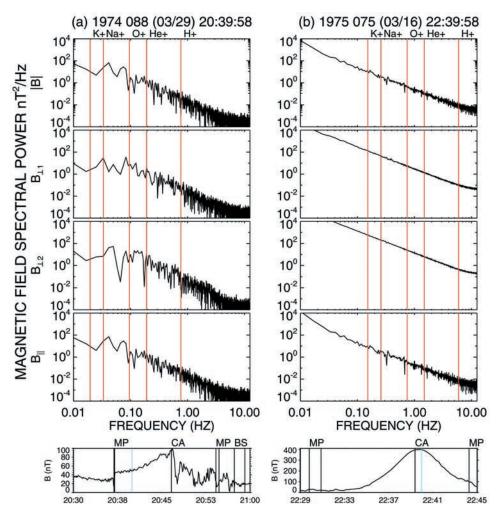


Figure 3. (a) A spectrum taken during Mariner 10's first Mercury flyby just inside the magnetopause. The spectral power is plotted for the magnitude, the transverse components, and the parallel components. The red vertical lines indicate the locations of the cyclotron frequencies of the indicated ion species. The lower panel is B magnitude plotted versus times during the flyby, and the blue vertical bar indicates where the spectrum was made. A spectral peak near $1.2 f_{cNa+}$ is observed in the parallel component and in one of the perpendicular components. The strong compressional component in the wave is not characteristic of an ICW. (b) Typical spectrum taken during the third flyby. No evidence of transverse wave activity near f_{cNa+} was observed during this flyby.

1974] and \sim 50 nT for B gives an V_A of 635 km/s. Since Mariner-10 was located in the lobe near the magnetopause, we assume that $V_{F\parallel}$ is 1/4 of the solar wind velocity of 660 km/s [*Ogilvie et al.*, 1974]. Using these values the frequency would be Doppler shifted by $0.6 \, f_{cNa+}$.

[10] During this time period, at higher frequencies (0.1 to 0.4 Hz), Russell [1989] showed that the compressional amplitude decreased as Mariner 10 moved away from the magnetopause suggesting a magnetopause source. We conclude that the spectral peak at 0.04 Hz could be explained by magnetopause motion. A one cycle undulation whose duration is near the Na⁺ cyclotron period could be due to many other factors other than Na⁺ cyclotron waves. Figure 2b shows an example spectrum made inside the magnetosphere during the third flyby, we could find no examples of dominant spectral peaks in the transverse components near f_{cNa^+} for this flyby.

[11] An example of a positive result would be O⁺ cyclotron waves observed in Saturn's middle magneto-

sphere by *Russell et al.* [2006]. Here, wave trains lasted hundreds of cycles, while the transverse spectral power of the spectral peak near the O⁺ cyclotron frequency was two orders of magnitude larger than the background and over one order of magnitude larger than the compressional power. Ideally one would want at least 5 wave periods [*Song and Russell*, 1999] in the wave amplitude time series to believe in the existence of a spectral peak. Thus Mariner 10's flybys are marginal due to their short dwell times in the regions of interest.

4. Discussion

[12] Because the size of Mercury's magnetosphere is small and the Larmor radii of Na⁺ is large, under what conditions will the ICW wavelength be large relative to typical scale sizes in Mercury's magnetosphere? To estimate these wavelengths we will use the approach developed by *Thorne and Tsurutani* [1987]. In their paper they investi-

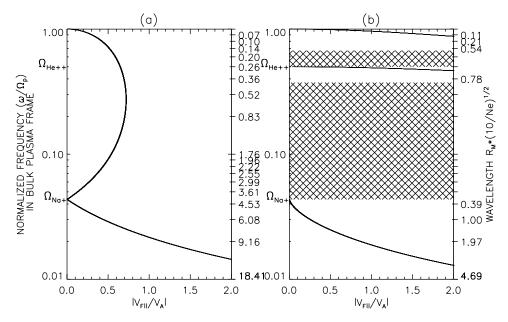


Figure 4. Frequency in the bulk plasma frame normalized by f_{cp} ($\omega/\Omega_{\rm p}$) plotted versus $V_{F\parallel}/V_A$ is shown for a bulk plasma composition of (a) 100% H⁺ and (b) 45% H⁺, 5% He⁺⁺, and 50% Na⁺. The magnetic field dependence is folded into the normalization. The wavelength in R_M for the corresponding frequency is given on the right axis and is computed for a plasma density N_e of 10 cm⁻³ and scales as $\sqrt{10/N_e}$. The cross hatched areas indicate that a real solution for the wavelength does not exist in that frequency range.

gated the generation of ICWs associated with H_2O^+ cometary pick-up ions in the solar wind. In our calculations H_2O^+ will be replaced by Na^+ . The wave mode is supported by the flowing (bulk) plasma and it must be in cyclotron resonance with the Na^+ in the pick-up frame. In general satisfying this resonance condition creates solutions whose frequency in the flow frame can be either lower or higher than f_{cNa^+} . The frequency in the flow frame determines the wavelength.

[13] First we look at the low frequency solutions in the bulk plasma frame in which the frequency is below the smallest ion cyclotron frequency of the ions that make up the bulk plasma (i. e. not the pickup ions). As explained by *Thorne and Tsurutani* [1987], these waves, which must always be left handed (L) in the pick-up ion frame, will be L-mode in the bulk flow frame if their phase velocity is greater than $V_{F\parallel}$, otherwise they will be R-mode. The low frequency solution yields wavelengths that can be approximated by the following.

$$\lambda_{\parallel} = (V_A/f_{cNa+}) \left(1 + \left| V_{F\parallel}/V_A \right| \right) \tag{1}$$

The length factor $V_A | f_{cNa^+}$ is $13.8 / \sqrt{\overline{\mu} N_e}$ in R_M , and $\overline{\mu}$ is the average mass density of the bulk plasma in *amu*. The factor $1 + |V_{F\parallel} V_A|$ arises from the Doppler shift of the frequency between the pick-up ion frame and the bulk plasma flow frame. In general these wavelengths are on the order of the size of Mercury's magnetosphere. Since the source region will be much smaller than these wavelengths, it is doubtful that they will grow to sufficient amplitudes to effectively scatter and thermalize Na^+ pick-up ions.

[14] Solutions for L-mode frequencies (in the bulk plasma flow frame) that are resonant with the Na⁺ pick-up ions versus $V_{F\parallel}/V_A$ are shown in Figures 4a and 4b, which are

analogous to Figure 5 of *Thorne and Tsurutani* [1987]. Because solutions are L-mode in the flow frame, the effects of ion cyclotron resonances for ion species that make up the bulk plasma become important in determining the wavelengths. Wavelength in $R_{\rm M}$ for the corresponding frequency is indicated on the right axis, and scale as $\sqrt{10/N_e}$. Looking at Figure 4a, one can see that the only way to get wavelengths sufficiently smaller than 1 $R_{\rm M}$ is for the wave frequency to be very close to the proton cyclotron frequency in the bulk plasma frame. For a multi-component plasma (Figure 4b) one can only get small wavelengths near an ion cyclotron resonance frequency of one of the ion species that make up the bulk plasma (i.e. not to be confused with the pickup ions).

[15] For a multi-component background plasma, the solution near each ion cyclotron resonance frequency (in the bulk plasma flow frame) can be approximated by the following.

$$\lambda_{\parallel} = \left(ca_{i}/f_{pi}\right) \frac{V_{F\parallel}/V_{A}}{1 - \left(a_{i}V_{F\parallel}/V_{A}\right)^{2}} \tag{2}$$

The length factor ca_i/f_{pi} is 0.6 $\mu_i/\sqrt{\mu N_e}$ in R_M , $a_i = \sqrt{\eta_i \mu_i/\overline{\mu}}$, c is the speed of light, f_{pi} is the ion plasma frequency, μ_i is the mass species i in amu and η_i is the fractional ion composition of species i. For the H^+ branch the error in this approximation is less than 10% of the true solution for $V_{F\parallel}/V_A < 0.6$. As $V_{F\parallel}/V_A$ approaches zero, the wave frequency approaches proton cyclotron resonance and the wavelength can be made arbitrarily small. However, as the frequency approaches this resonance, proton cyclotron damping of this wave will increase.

[16] In order to get sufficient wave growth these waves must be in Na⁺ cyclotron resonance as they propagate,

therefore, the effects of gradients in the magnetic field and plasma density are important in determining the size of the growth region. For simplicity we will use a dipole magnetic field model for computing the gradients. Assuming that the waves as they propagate are no longer in resonance and therefore cannot be further amplified once $\delta\omega$ / $\Omega_{Na+} = (\nabla B/B)L \approx (3/R_M)L > 0.1$, and that at least 10 wavelengths ($L = 10\lambda_{\parallel}$) are necessary for sufficient amplification, the upper limit on the wavelength can be estimated as:

$$\max \lambda_{\parallel} = 0.01B/\nabla B = 0.01R_M/3 \tag{3}$$

[17] From (2) and (3) an upper limit on the $V_{F\parallel}/V_A$ can be given by

$$V_{F\parallel}/V_A < 0.006R_M \sqrt{\overline{\mu}N_e}/\mu_i \tag{4}$$

- [18] This condition can be very restrictive regarding where these waves can be generated in Mercury's magnetosphere. Additionally, as noted, cyclotron wave damping by the lighter ions could be a problem. We conclude that thermalization of Na⁺ on a global scale by these waves is not possible, at best one can hope for is localized wave generation and thermalization in a small number of regions.
- [19] Based on the criteria given by (4) Na⁺ ICW generation is not likely in regions of low plasma density like the lobes. From Helios-1 observations (0.31–0.35 AU) the nominal solar wind density is about 64 cm⁻³, which gives an upper limit for the plasma sheet density of ~ 10 cm⁻³ [Mukai et al., 2004] near nominal conditions. From (4) this gives a value of $V_{F\parallel}/V_A < 0.018$ in the plasma sheet for conditions favorable to ICW generation. This criterion is very restrictive, but could be satisfied during dipolarizations.
- [20] The magnetosheath and magnetopause boundary layers are probably the best regions for generation of ICWs associated with Na⁺. Taking 4 times the nominal solar wind value as a density estimate in the day-side magnetosheath one gets from (4) $V_{F||}/V_A < 0.1$, this criteria is much more favorable for ICW generation in the nose of the magnetosheath. However, between 0.31-0.35 AU, IMF $|B_X|$ tends to be 2.5 times larger than IMF $|B_Y|$ or $|B_Z|$, and therefore magnetic field direction tends to be aligned with plasma flow in the flanks of the magnetosheath. The generation of Na⁺ ICW in the flanks should be a rare occurrence. Only in the subsolar magnetosheath can the flow be close to perpendicular to the field direction. Additionally conditions are more favorable for $V_{F\parallel} \leq V_A$ in this region. Therefore, in the subsolar magnetosheath it might be possible to generate ICWs of sufficient amplitude to thermalize the pick-up Na⁺ ions and a good fraction of these ions could enter the magnetosphere through the cusps. To investigate this possibility hybrid simulations [i.e., Omidi et al., 2006; Travnicek et al., 2007] need to be modified to include heavy ions.

5. Conclusion

[21] Due to the strong overlap between Mercury's Na exosphere and its outer magnetosphere and magnetosheath one would expect a wave signature as newly created Na⁺

- ions are "picked-up". The magnetometer data from the first and third Mariner 10 flybys were examined for the evidence of Na⁺ ICW expected to be produced by the thermalization process. However, no evidence of Na⁺ ICW was found. There are several possible reasons for the negative result. One is that the freshly created Na⁺ pick-up ions are lost from the Mercury system due to their large Larmor radii before there is time for the ICWs to grow to detectable amplitudes. Another is the very brief nature of Mariner 10's transversal through Mercury's magnetosheath and magnetosphere, about 15 min in total. A general rule is that, at least 5 wave oscillations are necessary for the determination of a spectral peak. Our calculations indicate that Mariner 10 would only have been present in each of the key regions about Mercury for a maximum of ~3–4 ICW wave periods.
- [22] Furthermore, we performed additional calculations which indicate that the wavelengths of Na⁺ ICWs and those associated with other heavy ions are on the order of the system scale lengths. It is, therefore, questionable whether such long wavelength waves can grow to sufficient amplitudes to effectively thermalize these planetary pick-up ions in the magnetosheath or magnetosphere.
- [23] In summary, our examination of the Mariner 10 measurements and simple estimates of heavy ion ICW properties indicate that such waves are not only difficult to detect during brief flybys, but the small dimensions of Mercury's magnetosphere may greatly constrain their growth. If the small size of Mercury's magnetosphere does inhibit ICW wave growth, then the ability of heavy planetary ions to be assimilated and play a significant role in the dynamics of Mercury's magnetosphere may be in doubt.
- [24] Clearly more data is needed from a spacecraft with much longer dwell times. For this we will have to wait for the MESSENGER and BepiColombo missions to reach Mercury. The dwell times by MESSENGER in Mercury's magnetosphere during its 3 flybys will be ~ 1.6 times larger than that of Mariner, and once in orbit MESSENGER's dwell time in key regions will be 10 to 100 times larger.
- [25] **Acknowledgments.** We acknowledge the PDS (http://pds.nasa.gov/) and COHOWEB for providing data. We thank R. Lepping for discussions about Mariner 10. This work was supported by NASA's MESSENGER Mission.

References

Cheng, A. F., R. E. Johnson, S. M. Krimigis, and L. J. Lanzerotii (1987), Magnetosphere, exosphere, and surface of Mercury, *Icarus*, 71, 430–440

Delcourt, D. C., T. E. Moore, S. Orsini, A. Millilo, and J. A. Sauvaud (2002), Centrifugal acceleration of ions near Mercury, *Geophys. Res. Lett.*, 29(12), 1591, doi:10.1029/2001GL013829.

Delcourt, D. C., S. Grimald, F. Leblanc, J.-J. Berthelier, A. Millilo, A. Mura, S. Orsini, and T. E. Moore (2003), A quantitative model of planetary Na⁺ contribution to Mercury's magnetosphere, *Ann. Geophys.*, 21, 1723–1736.
Ip, W.-H. (1986), The sodium exosphere and magnetosphere of mercury, *Geophys. Res. Lett.*, 13, 423–426.

Lee, M. A. (1989), Ultra-low frequency waves at comets, in *Plasma Waves* and *Instabilities at Comets and in Magnetospheres, Geophys. Monogr. Ser.*, vol. 53, edited by B. T. Tsurutani and H. Oya, pp. 13–29, AGU, Washington, D. C.

Lepping, R. P., N. F. Ness, and K. W. Behannon (1979), Summary of Mariner 10 magnetic field and trajectory data for Mercury I and III encounters, *NASA Tech. Memo.*, 80600, NASA Goddard Flight Cent., Greenbelt, Md.

Mukai, T., K. Ogasawara, and Y. Saito (2004), An empirical model of the plasma environment around Mercury, *Adv. Space Res.*, *33*(12), 2166–2171.

- Ogilvie, K. W., J. D. Scudder, R. E. Hartle, G. L. Siscoe, H. S. Bridge, A. J. Lazarus, J. R. Asbridge, S. J. Bame, and C. M. Yeates (1974), Observations at Mercury encounter by the plasma science experiment on Mariner 10, Science, 185(4146), 145-151.
- Omidi, N., X. Blanco-Cano, C. T. Russell, and H. Karimabadia (2006), Global hybrid simulations of solar wind interaction with Mercury: Magnetospheric boundaries, Adv. Space Res., 38(4), 632-638.
- Othmer, C., K.-H. Glassmeirer, and R. Cramm (1999), Concerning field line resonances in Mercury's magnetosphere, J. Geophys. Res., 104, 10,369 - 10,378.
- Potter, A. E., and T. H. Morgan (1985), Discovery of sodium in the atmosphere of Mercury, Science, 229, 651-653.
- Potter, A. E., and T. H. Morgan (1997), Evidence for suprathermal sodium on Mercury, Adv. Space Res., 19, 1571-1576.
- Potter, A. E., R. M. Killen, and T. H. Morgan (2002), The sodium tail of Mercury, Meteorit. Planet. Sci., 37, 1165-1172.
- Russell, C. T. (1989), ULF waves in the Mercury magnetosphere, Geophys.
- Res. Lett, 16, 1253–1256.
 Russell, C. T., J. S. Leisner, C. S. Arridge, M. K. Dougherty, and X. Blanco-Cano (2006), Nature of magnetic fluctuations in Saturn's middle magnetosphere, J. Geophys. Res., 111, A12205, doi:10.1029/ 2006JA011921
- Slavin, J. A. (2004), Mercury's magnetosphere, Adv. Space Res., 33(11), 1859-1874.

- Song, P., and C. T. Russell (1999), Time series data analyses in space physics, Space Sci. Rev., 87(3-4), 387-463.
- Terasawa, T. (1989), Particle scattering and acceleration in a turbulent plasma around comets, in Plasma Waves and Instabilities at Comets and in Magnetospheres, Geophys. Monogr. Ser., vol. 53, edited by B.T. Tsurutani and H. Oya, pp. 41-49, AGU, Washington, D. C.
- Thorne, R. M., and B. T. Tsurutani (1987), Resonant interactions between cometary ions and low-frequency electromagnetic waves, Planet. Space. Sci., 35(12), 1501-1511.
- Travnicek, P., P. Hellinger, and D. Schriver (2007), Structure of Mercury's magnetosphere for different pressure of the solar wind: Three dimensional hybrid simulations, Geophys. Res. Lett., 34, L05104, doi:10.1029/ 2006GL028518.
- Tsurutani, B. T. (1991), Cometary plasma waves and instabilities, in *Comets* in the Post-Halley Era, vol. 2, edited by R. L. Newburn Jr. et al., pp. 1171-1210, Kluwer Acad., Dordrecht, Netherlands.
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